Int. Appl. No.: PCT/US02/15387 U.S. Appl. No.: 10/706,925

Amdt. dated: July 19, 2005

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Amendments to the Specification:

Please amend paragraph [0009] as shown in the following amended paragraph:

For the coolant, this commonly flows across the back of each fuel cell, so [0009]as to flow between adjacent, individual fuel cells. This is not essential however and, as a result, many fuel cell stack designs have cooling channels only at every 2nd, 3rd or 4th (etc.) plate. This allows for a more compact stack (thinner plates) but may provide less than satisfactory cooling. This provides the requirement for another seal, namely a seal between each adjacent pair of individual fuel cells. Thus, in a completed fuel cell stack, each individual fuel cell will require two seals just to seal the membrane exchange electrode assembly to the two flow field plates. A fuel cell stack with 30 individual fuel cells will require 60 seals just for this purpose. Additionally, as noted, a seal is required between each adjacent pair of fuel cells and end seals to current collectors. For a 30 cell stack, this requires an additional 31 seals. Thus, a 30 cell stack would require a total of 91 seals (excluding seals for the bus bars, current collectors insulator plates and endplates), and each of these would be of a complex and elaborate construction. With the additional gaskets required for the bus bars, insulator plates and endplates the number reaches 100 seals, of various configurations, in a single 30 cell stack.

Please amend paragraph [0014] as shown in the following amended paragraph:

[0014] A fuel cell stack, after assembly, is commonly clamped to secure the elements and ensure that adequate compression is applied to the seals and active area of the fuel cell stack. This method ensures that the contact resistance is minimized and the electrical resistance of the cells are at a minimum. To this end, a fuel cell stack typically has two substantial end plates, which are configured to be sufficiently rigid so that their deflection under pressure is within acceptable tolerances. The fuel cell also typically has current bus bars to collect and concentrate the current from the fuel cell to a small pick up point and the current is then transferred to the load via conductors. Insulation plates may also be used to isolate, both thermally and electrically, the current bus bars and endplates from each other. A plurality of Int. Appl. No.: PCT/US02/15387 U.S. Appl. No.: 10/706,925

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elongated rods, bolts and the like are then provided between the pairs of plates, so that the fuel cell stack can be clamped together between the plates, by the tension rods. Rivets, straps, piano wire, metal plates and other mechanisms can also be used to clamp the stack together. To assemble the stack, the rods are provided extending through one of the end plates. aAn insulator plate and then a bus bar (including seals) are placed on top of the endplate, and the individual elements of the fuel cell are then built up within the space defined by the rods or defined by some other positioning tool. This typically requires, for each fuel cell, the following steps:

- (a) placing a seal to separate the fuel cell from the preceding fuel cell;
- (b) locating a flow field plate on the seal;
- (c) locating a seal on the first flow field plate;
- (d) placing a GDM within the seal on the flow field plate;
- (e) locating a membrane electrode assembly (MEA) on the seal;
- (f) placing an additional GDM on top of the MEA;
- (g) preparing a further flow field plate with a seal and placing this on top of the membrane exchangeelectrode assembly, while ensuring the seal of the second plate falls around the second GDM;
- (h) this second or upper flow field plate then showing a groove for receiving a seal, as in step (a).

Please amend paragraph [0015] as shown in the following amended paragraph:

[0015] This process needs to be <u>completed repeated</u> until the last cell is formed and it is then topped off with a bus bar, insulator plate and the final end plate.

Please amend paragraph [0021] as shown in the following amended paragraph:

[0021] In accordance with a first aspect of the present invention, there is provided a fuel cell assembly comprising:

a plurality of separate elements;

at least one groove network extended throughout the fuel cell assembly and including at least one filling port for the <u>at least one</u> groove network; and

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a seal within each groove network that has been formed in place after assembly of said separate elements, wherein the seal provides a seal between at least two of said separate elements to define a chamber for a fluid for operation of the fuel cell.

Please amend paragraph [0028] as shown in the following amended paragraph:

In addition, the ability of the seal to bond the elements together facilitates the production of membrane electrode units (MEU). As detailed below, such membrane electrode units could each comprise a single fuel cell or a small number of fuel cells. Each unit would have end surfaces adapted for mating within surfaces of corresponding membrane electrode units, e.g. to form coolant chambers; for this purpose, a seal can be molded on one or both ends of each membrane electrode unit. The membrane electrode units can then be assembled and clamped together to form a fuel cell stack of a desired power level.

Please amend paragraph [0033] as shown in the following amended paragraph:

The first embodiment of the apparatus is shown in Figure 1a and indicated generally by the reference 20. For simplicity, this Figure shows just part of a fuel cell stack, as does Figure 2. It will be understood that the other fuel cells in the stack correspond, and that the fuel cell stack would include conventional end elements, clamping elements and the like. In general, Figures 1a-3 are intended to indicate the essential elements of the individual embodiments of the invention, and it will be understood by someone skilled in this art that the fuel cell stacks would be otherwise conventional. Also in Figures 1a-e and 2, the proton exchange member membrane is shown, for clarity, with exaggerated thickness, and as is known, it has a small thickness. In Figures 1a-e, the grooves for the seal material are shown schematically, and it is expected that the grooves will usually have a depth and width that are similar, i.e. a generally square cross-section. Note also that the bottom of the grooves can have any desired profile.

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Please amend paragraph [0035] as shown in the following amended paragraph:

[0035] As isn conventional, the MEA is considered to comprise a total of three layers, namely: a central proton exchange membrane layer (PEM); on both sides of the PEM, a layer of a finely divided catalyst, to promote reaction necessary on either side of the PEM. There are also two layers of gas diffusion media (GDM) located on either side of the PEM abutting the catalyst layers, and usually maintained pressed against the catalyst layers to ensure adequate electrical conductivity, but these two layers of GDM are not considered to be part of the MEA itself.

Please amend paragraph [0040] as shown in the following amended paragraph:

[0040] Figure 2 shows an alternative arrangement. Here, the anode and cathode bipolar plates are indicated at 42, 44 and 42a, corresponding to plates 22 and, 24 and 22a of Figure 1a. The MEA is again indicated at 26. A coolant cavity is formed at 46, and cavities or chambers 48, 50 are provided for the GDM.

Please amend paragraph [0044] as shown in the following amended paragraph:

Figure 3 shows an assembly device indicated generally at 60, for forming a seal, somewhat as for the embodiment of Figure 2. Here, it is anticipated that a fuel cell stack will first be assembled following known practice, but without inserting any seals. Thus, the various elements of the stack, principally the flow fuelfield plates and the MEAs will be sequentially assembled with appropriate end components. To align the components, clamping rods can be used by first attaching these to one end plate, or the components can be assembled in a jig dimensioned to ensure accurate alignment. Either way, with all the components in place the entire assembly is clamped together, commonly by using clamping rods, as mentioned, engaging both end plates. The assembly device 60 has a base 62 and a peripheral wall 64 defining a well 66. Additionally, there are upper and lower projections 68, for engaging the end plates to locate a fuel cell stack in position. Although Figure 3b shows the projections 68 on just two sides of the fuel cell stack, it will be understood that they are provided on all four sides.

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Please amend paragraph [0053] as shown in the following amended paragraph:

[0053] Referring first to Figures 5 and 6, there are shown the basic elements of the stack 100. Thus, the stack 100 includes an anode endplate 102 and cathode endplate 104. In known manner, the endplates 102, 104 are provided with connection ports for supply of the necessary fluids. Air connection ports are indicated at 106, 107; coolant connection ports are indicated at 108, 109; and hydrogen connection ports are indicated at 110, 111. Although not shown, it will be understood that corresponding air, coolant and hydrogen ports, corresponding to ports 106-111 would be provided on the anode side of the fuel cell stack. The various ports 106-111 are connected to distribution channels or ducts that extend through the fuel cell stack 100, as for the earlier embodiments. The ports are provided in pairs and extend all the way through the fuel cell stack 100, to enable connection of the fuel cell stacks to be connected together, in known manner.

Please amend paragraph [0058] as shown in the following amended paragraph:

[0058] Reference will now be made to Figures 6a and 6b, which show configurations with respectively, 20 and 100 individual fuel cells. These Figures show the fuel cells schematically, and indicate the basic elements of the fuel cells themselves, without the components necessary at the end of the stack. Thus, endplates 102, 104, insulators 112, 114, and current collectors 106116, 108118 are not shown. Instead, these Figures simply show pairs of flow field plates 120, 130.

Please amend paragraph [0059] as shown in the following amended paragraph:

[0059] In the following description, it is also to be understood that the designations "front" and "rear" with respect to the anode and cathode flow field plates 120, 130, indicates their orientation with respect to the MEA. Thus, "front" indicates the face towards the MEA; "rear" indicates the face away from the MEA. Consequently, in Figures 98 and 10, the configuration of the ports is reversed as compared to Figures 7 and 89.

Please amend paragraph [0066] as shown in the following amended paragraph:

[0066] Rectangular groove portion 144, for the <u>air flow aperture</u> 136, includes outer groove segments 148, which continue into a groove segment 149, all of which have a width of 0.200". An inner groove segment 150 has a width of 0.120". For the aperture 138 for cooling fluid, a rectangular groove 145 has groove segments 152 provided around three sides, each again having a width of 0.200". For the aperture 140, a rectangular groove 146 has groove segments 154 essentially corresponding with the groove segments 152 and each again has a width of 0.200". For the groove segments 152, 154, there are inner groove segments 153, 155, which like the groove segment 150 have a width of 0.120".

Please amend paragraph [0067] as shown in the following amended paragraph:

[0067] It is to be noted that, between adjacent pairs of apertures 136, 138 and 138, 140, there are groove junction portions 158, 159 having a total width of 0.5", to provide a smooth transition between adjacent groove segments. This configuration of the groove junction portions 158, 159 and the reduced thickness of the groove segments 150, 153, 155, as compared to the outer groove segments, is intended to ensure that the material for the sealant flows through all the groove segments and fills them uniformly.

Please amend paragraph [0068] as shown in the following amended paragraph:

[0068] To provide a connection through the various flow field plates and the like, a connection aperture 160 is provided, which has a width of 0.25", rounded ends with a radius of 0.125" and an overall length of 0.35". As shown, in Figure 7 connection aperture 160 is dimensioned so as to clearly intercept the groove segments 152, 154. This configuration is also found in the end plates, insulators and current collection plates, as the connection aperture 160 continues through to the end plates and the end plates have a corresponding groove profile. It is seen in greater detail in Figures 12 and 15, and is described below.

Please amend paragraph [0077] as shown in the following amended paragraph:

Thus, for the anode end plate 102, there is a groove network 190, that corresponds to the groove network on the <u>front rear</u> face of the anode flow field plate 120. Accordingly, similar reference numerals are used to designate the different groove segments of the anode and <u>anode cathode</u> end plates 102, 104 shown in detail in Figures 11-13 and 14-15, but identified by the suffix "e". As indicated at 192, threaded bores are provided for receiving the tie rods <u>132131</u>.

Please amend paragraph [0088] as shown in the following amended paragraph:

[0088] The two flows around the aperture 140 will eventually lead into the side groove 143. It will be appreciated that the dimensions of the grooves 154, 155 and the location of the connection aperture 160 are chosen such that the two fields flows will meet approximately simultaneously, and in particular, that no air pocket will be left.

Please amend paragraph [0089] as shown in the following amended paragraph:

[0089] Correspondingly, the flows around the aperture 138 will meet at the groove junction portion 158. Again, the dimensions of the groove segments 152, 153 and also the groove junction portion 159 are sized to ensure that these flows meet approximately simultaneously. The flow then diverges again and flows in two paths around the larger aperture 136 for the oxidant flow. Note that again the groove segment 148 has a larger width than the groove segment 149150, to promote approximately equal travel time around the aperture 136, so that the two flows arrive generally simultaneously at a junction with the topmost groove 143 in Figure 7. The flows then combine to pass down the side groove 143.

Please amend paragraph [0092] as shown in the following amended paragraph:

[0092] In practice, for any particular fuel stack assembly, tests will be run to establish the filling time required to ensure complete filling of all grooves and channels. This can be done for different materials, dimensions, temperatures etc. With the filling time established, material is then injected into the complete stack assembly 100, for the

determined filling time, following which the flow is terminated, and the seal material supply is detached.

Please amend paragraph [0095] as shown in the following amended paragraph:

[0095] If any leaks are detected, the fuel cell will most likely have to be repaired. Depending on the nature of the leak and details of an individual stack design, it may be possible simply to separate the whole assembly at one seal, clear out the defective seal and then form a new seal. For this reason, it may prove desirable to manufacture relatively small fuel cells stacks, as compared to other conventional practice. While this may require more inter-stack connections, it will be more than made up for by the inherent robustness of and reliability of each individual fuel cell stack. The concept can be applied all the way down to a single cell unit (identified as a Membrane Electrode Unit or MEU) and this would then conceivably allow for stacks of any length to be manufactured.

Please amend paragraph [0096] as shown in the following amended paragraph:

This MEU is preferably formed so that a number of such MEU's to can [0096] be readily and simply clamped together to form a complete fuel cell stack of desired capacity. Thus, an MEU would simply have two flow field plates, whose outer or rear faces are adapted to mate with corresponding faces of other MEU's, to provide the necessary functionality. Typically, faces of the MEU are adapted to form a coolant chamber of for cooling fuel cells. One outer face of the MEU can have a seal or gasket preformed with it. The other face could then be planar, or could be grooved to receive the preform seal on the other MEU. This outer seal or gasket is preferably formed simultaneously with the formation of the internal seal, injected-in-place in accordance with the present invention. For this purpose, a mold half can be brought up against the outer face of the MEU, and seal material can then be injected into a seal profile defined between the mold half and that outer face of the MEU, at the same time as the seal material is injected into the groove network within the MEU itself. To form a complete fuel cell assembly, it is simply a matter of selecting the desired number of MEU's, clamping the MEU's together between endplates, with usual additional end

components, e.g. insulators, current collectors, etc. The outer faces of the MEU's and the preformed seals will form necessary additional chambers, especially chambers for coolant, which will be connected to appropriate coolant ports and channels within the entire assembly. This will enable a wide variety of fuel cell stacks to be configured from a single basic unit, identified as an MEU. It is noted, the MEU could have just a single cell, or could be a very small number of fuel cells, e.g. 5. In the completed fuel cell stack, replacing a failed MEU, is simple. Reassembly only requires ensuring that proper seals are formed between adjacent MEU's and seals within each MEU are not disrupted by this procedure.

Please amend paragraph [0098] as shown in the following amended paragraph:

[0098] A second consideration is that providing the MEA, or a flange layer, bisecting a groove or channel for the seal material may give problems. It is assumed that flow of the seal material is uniform. This may not occur in practice. For example, if the MEA distorts slightly, then flow cross-sections on either side will distort. This will lead to distortions in flow rates of the seal material on the two sides of the MEA, which will only cause the distortion to increase. Thus, this will increase the flow on the side already experiencing greater flow, and restrict it on the other side. This can result in improper sealing of the MEA. To avoid this, the invention also anticipates variants, shown in Figures 1b-1e. These are described below, and for simplicity like components in these figures are given the same reference numerals as in Figure 1a, but with the suffixes b,c,d are as appropriate, to indicate features that are different.

Please amend paragraph [0103] as shown in the following amended paragraph:

[0103] In Figure 1d and 1e, the anode and cathode flow field plates have flat, opposing faces, although it will be understood that these faces, in known manner, would include flow channels for gases. As these faces are otherwise flat, this greatly eases tolerance and alignment concerns, and in general it is expected that the MEA 26d,e can be inserted without requiring en any tight tolerances to be maintained.

Please amend paragraph [0118] as shown in the following amended paragraph:

[0118] In these Figures 17-20, the transfer slots are denoted by the references 178a, for the anode plate 120, and 180a, for the cathode plate 130. The suffixes indicate that the transfer slots have different dimensions and more numerous. There are eight transfer slots 178a, as compared to four slots 178, and there are either eight (Figure 19) or ten (Figure 20) transfer slots 180a, as compared to five slots 180. It will also be understood that it is not necessary to provide discrete slots and that, for each flow, it is possible to provide a single relatively large transfer slot. Each of the slots 178a communicates with a single flow channel (Figure 17), and each fo the slots 180a communicates with two flow channels, except for an end slot 180a that communicates with a single channel (Figure 19).

Please amend paragraph [0122] as shown in the following amended paragraph:

Preferably, the polydiorganosiloxane has a viscosity within a range of about 0.03 to less than 100 Pa·s at 25e°C. The polydiorganosiloxane can be represented by the general formula X(R1R2SiO)nX where R1 and R2 represent identical or different monovalent substituted or unsubstituted hydrocarbon radicals, the average number of repeating units in the polymer, represented by n, is selected to provide the desired viscosity, and the terminal group X represents an ethylenically unsaturated hydrocarbon radical. For example, when the composition is to be cured by a hydrosilylation reaction with an organohydrogensiloxane or a vinyl-specific peroxide, X is typically vinyl or other alkenyl radical.

Please amend paragraph [0126] as shown in the following amended paragraph:

The type of reinforcing silica filler used in the present process is not critical and can be any of those reinforcing silica filler known in the art. The reinforcing silica filler can be, for example, a precipitated or pyrogenic silica having a surface area of at least 50 square meters per gram (M2/g). More preferred is when the reinforcing silica filler is a precipitated or pyrogenic silica having a surface area within a range of about 150 to 500 M2/g. The most preferred reinforcing silica filler is a pyrogenic silica having a surface area of about 370 to 420 M2/g. The pyrogenic silica filler can be

produced by burning silanes, for example, silicon tetrachloride or trichlorosilane as taught by Spialter et al. U.S. Pat. No. 2,614,906 and Hugh et al. U.S. Pat. No. 3,043,660. The aforementioned fillers can be treated with a silazane, such as hexamethyldisilazane, an organosilane, organopolysiloxane, or other organic silicon compound. The amount of this ingredient added depends on the type of the inorganic filler used. Usually, the amount of this ingredient is in the range of 5 to 50 parts by weight per 100 parts by weight of ingredient (ba).

Ingredient (c), (Oxide or Hydroxide of an Alkaline-Earth Metal)

Please amend paragraph [0138] as shown in the following amended paragraph:

[0138] 100 parts of silicone base material (as mentioned in Table 1 above); 56 parts dimethylpolysiloxane that is dimethylvinylsiloxy terminated on both ends and has a viscosity of 55,000 cp; 34 parts dimethyl, methylvinylsiloxane which is dimethylvinylsiloxy terminated and has a viscosity of 350 cp; 12 parts of calcium hydroxide which is certified 99% pure and contains a sulfur content of less thenthan 0. 1%; and 0.7 parts of 1,3-diethenyl-1,1,3,3-tetramethyldisiloxane platinum complexes which contains an amount of platinum metal atoms equaling 0.52 wt % were blended until homogeneity. Final material is a flowable liquid silicone with a viscosity of 128,000 cp at 23 °C.

Please amend paragraph [0140] as shown in the following amended paragraph:

[0140] The final compositions of material A and material B from above when mixed in a 50:50 ratio and press molded at 150<u>°</u>C for 5 minutes exhibit the following characteristics:

Please amend paragraph [0144] as shown in the following amended paragraph:

[0144] A stack of fuel cell elements was assembled using the following procedure (with reference to the structure of Figure 5): 1), place an aluminum anode end plate 102 flat on a horizontal surface, with the seal groove segments facing up; 2), place a high-density polyethylene insulator plate 112 on the anode end plate, locating

the plate so the seal groove segments on each plate align with each other; 3), place a gold-plated nickel anode bus bar plate 116 on the insulator plate, locating the plate so the seal groove segments on each plate align with each other; 4), place an anode bipolar flow field plate 120 on the insulator plate with the active area facing up, aligning the groove segments and apertures of each plate; 5), place a GDML ply 122, cut to fit in the recessed surface active area of the anode bipolar flow field plate; 6), place a PEM ply 124 on the anode bipolar flow field plate and GDML, making sure that the apertures for flowing seal material are aligned with the aperture on the flow field plate; 7), place a GDML ply 126, cut to fit in the recessed surface active area of the cathode bipolar flow field plate; 8), place a cathode bipolar flow field plate 130 on the assembly, with the active area facing down; 9), place a gold-plated nickel cathode bus bar plate 118 on the assembly, locating the plate so the seal groove segments and apertures align; 10), place a high-density polyethylene insulator plate 114 on the assembly, locating the plate so the seal groove segments and the apertures on each plate align with each other; 11), place the aluminum cathode end plate 104 flat on a horizontal surfaceon the assembly, with the seal groove segments facing down; 12), place perimeter bolts or tie rods 131 through the cathode end plate 104 that extend to screw into the anode end plate 102; 13), tighten the perimeter bolts 131 to provide even clamping of the assembly elements, items 1) through 11).

Please amend paragraph [0149] as shown in the following amended paragraph:

[0149] 100 parts of a polydimethylsiloxane_which is dimethylvinylsiloxy terminated, is 30 mole % methyltrifluoropropyl, and had a viscosity of 9,300 cst; 1 part of dimethylmethylvinylsiloxane which is hydroxy terminated and had a viscosity of 40 cst; and 38 parts of fumed silica (with an average surface area of 250 m2/g) that had been surface-treated with 6.4 parts hexamethyldisilazane were blended until homogeneity was achieved. After blending, the material was heat treated under vacuum to drive off volatiles, so as to form a base material. This was then cut back or diluted with 10 parts of polydimethylsiloxane which is dimethylvinylsiloxy terminated, is 30 mole % methyltrifluoropropyl, and had a viscosity of 680 cst; 3_8 parts of dimethyl, hydrogensiloxy – modified silica with 0.96 wt % silicone-atom-bonded hydrogen atoms

and a viscosity of 25 cp; and 0.2 parts 1-ethynyl-1-cyclohexanol which was 99 % pure, for use as an inhibitor to the mixed system. The complete composition was blended until homogeneity. The final material or composition was a flowable silicone paste that could be extruded through an 1/8" orifice at a rate of 259.5 g/min under 90 psig pressure.

Please amend paragraph [0152] as shown in the following amended paragraph:

terminated, is 40 mole% methyltrifluoropropyl, and had a viscosity of 25,000 cst; 1 part of dimethylmethylvinylsiloxane which is hydroxy terminated and had a viscosity of 40 cst; and 38 parts of fumed silica (with an average surface area of 250 m2/g) that had been surface-treated with 6.4 parts hexamethyldisilazane and were blended until homogeneity was achieved. After blending, the material was heat treated to remove volatiles, so as to form a base material. This was then cut back or diluted with 10 parts of polydimethylsiloxane which is dimethylsiloxy terminated, is 40 mole % methyltrifluoropropyl, and had a viscosity of 750 cst; 3_8 parts of dimethyl, hydrogensiloxy — modified silica with 0.96 wt % silicone-atom-bonded hydrogen atoms and a viscosity of 25 cp; and 0.2 parts 1-ethynyl-1-cyclohexanol which was 99 % pure for use as an inhibitor to the mixed system. The complete composition was blended until homogeneity. The final material was a flowable silicone paste that could be extruded through an 1/8" orifice at a rate of 225 g/min under 90 psig pressure.

Please amend paragraph [0155] as shown in the following amended paragraph:

[0155] Several methods were used to determine resistance to the PEM operating environment. For example, sheets of seal material were placed in contact with sheets of PEM material, rolled tightly and held in position with appropriate banding. Such rolls were then placed in acidic fluids and, separately, heated DI water to provide an accelerated aging test. Such a test was completed with DI water heated to 100 degrees <u>°C</u> for the seal materials listed previously. After 1 month of exposure the material was not hardened or cracked.

Please amend paragraph [0157] as shown in the following amended paragraph:

100 parts of a dimethylsiloxane which is dimethylvinylsiloxy terminated, [0157] is 70 mole % methyltrifluoropropyl, and had a viscosity of 20,000 cst; 1 part of dimethylmethylvinylsiloxane which is hydroxy terminated and had a viscosity of 40 cst; and 38 parts of fumed silica (with an average surface area of 250 m2/q) that had been surface-treated with 6.4 parts hexamethyldisilazane and were blended until homogeneity was achieved. After blending, the material was heat treated under vacuum, to remove volatiles, so as to form a base material. This was then cut back or diluted with 10 parts of the polydimethylsiloxane which is dimethylvinylsiloxy terminated, is 70 mole % methyltrifluoropropyl, and had a viscosity of 1500 cst; 3.8 parts of dimethyl, hydrogensiloxy - modified silica with 0.96 wt % silicone-atombonded hydrogen atoms and a viscosity of 25 cp; and 0.2 parts 1-ethynyl-1cyclohexanol which was 99 % pure for use as an inhibitor to the mixed system. The complete composition was blended until homogeneity. The final material was a flowable silicone paste that could be extruded through an 1/8" orifice at a rate of (189) g/min under 90 psig pressure.